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WO 01/18846 A2

(51) International Patent Classification[7]: H01J 49/40

(21) International Application Number: PCT/GB00/03332

(22) International Filing Date: 31 August 2000 (31.08.2000)

(25) Filing Language: English

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(30) Priority Data:
9920711.0 3 September 1999 (03.09.1999) GB

(71) Applicant *(for all designated States except US)*: **MASSLAB LIMITED** [GB/GB]; Crewe Road, Wythenshawe, Manchester M23 9BE (GB).

(72) Inventors; and
(75) Inventors/Applicants *(for US only)*: **DAVIS, Stephen** [AU/GB]; 8 Flash Lane, Bollington, Macclesfield, Cheshire SK10 5AQ (GB). **MAKAROV, Alexander** [RU/GB]; 80 Marlborough Avenue, Cheadle Hulme, Cheadle, Cheshire SK8 7AR (GB). **HUGHES, Jonathan** [GB/GB]; Plum Cottage, Snelson Lane, Snelson, Macclesfield, Cheshire SK11 9BP (GB).

(74) Agents: **HILL, Richard** et al.; Wilson Gunn M'Caw, 41-51 Royal Exchange, Cross Street, Manchester M2 7BD (GB).

(81) Designated States *(national)*: CA, JP, US.

(84) Designated States *(regional)*: European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).

Published:
— *Without international search report and to be republished upon receipt of that report.*

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

WO 01/18846 A2

(54) Title: HIGH DYNAMIC RANGE MASS SPECTROMETER

(57) Abstract: A mass spectrometer comprises an ion source which produces an ion beam from a substance to be analysed and a detector to detect a quantity of ions incident thereon. The detector includes two elements (16, 18) each of which detect a part of the quantity of ions and an attenuation device attenuates the quantity of ions reaching one of the detector elements. At least one of the detector elements (16, 18) is connected to a time to digital converter (TDC) to allow counting of the ions and at least one of the detector elements is connected in parallel to both a time to digital converter (TDC) and an analogue to digital converter (ADC).

PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference	**FOR FURTHER ACTION** see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
RH/P/15339WO		
International application No.	International filing date *(day/month/year)*	(Earliest) Priority Date *(day/month/year)*
PCT/GB 00/ 03332	31/08/2000	03/09/1999
Applicant		
MASSLAB LIMITED		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 2 sheets.

[X] It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**

a. With regard to the **language,** the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

[ ] the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing :

[ ] contained in the international application in written form.

[ ] filed together with the international application in computer readable form.

[ ] furnished subsequently to this Authority in written form.

[ ] furnished subsequently to this Authority in computer readble form.

[ ] the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

[ ] the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. [ ] **Certain claims were found unsearchable** (See Box I).

3. [ ] **Unity of invention is lacking** (see Box II).

4. With regard to the **title,**

[X] the text is approved as submitted by the applicant.

[ ] the text has been established by this Authority to read as follows:

5. With regard to the **abstract,**

[X] the text is approved as submitted by the applicant.

[ ] the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No. 1

[X] as suggested by the applicant.

[ ] None of the figures.

[ ] because the applicant failed to suggest a figure.

[ ] because this figure better characterizes the invention.

INTERNATIONAL SEARCH REPORT

International Application No
PC[redacted] 00/03332

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 H01J49/40 H01J49/02 G01T1/28

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 H01J G01T

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
EPO-Internal, WPI Data, PAJ, INSPEC

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 99 38190 A (BATEMAN ROBERT HAROLD ;COTTRELL JONATHAN CHARLES (GB); GILBERT ANT) 29 July 1999 (1999-07-29)	1,3-6,8, 11-13
A	page 15 -page 16; figures 2-4	2,7,9
A	WO 98 21742 A (DAVIS LARRY J ;ROCKWOOD ALAN (US); SENSAR CORP (US)) 22 May 1998 (1998-05-22) cited in the application abstract	1

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
25 July 2001	01/08/2001

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31–70) 340–2040, Tx. 31 651 epo nl, Fax: (+31–70) 340–3016	Hulne, S

1

Form PCT/ISA/210 (second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
PCT 00/03332

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 9938190 A	29-07-1999	EP 0970504 A	12-01-2000
		EP 0970505 A	12-01-2000
		EP 0970506 A	12-01-2000
		WO 9938191 A	29-07-1999
		WO 9938192 A	29-07-1999
		JP 2001503196 T	06-03-2001
		JP 2000513494 T	10-10-2000
		US 6229142 B	08-05-2001
WO 9821742 A	22-05-1998	US 5777326 A	07-07-1998
		US 6163032 A	19-12-2000
		AU 7181898 A	03-06-1998
		EP 0939970 A	08-09-1999

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(71) Applicant *(for all designated States except US)*: **MASS-LAB LIMITED** [GB/GB]; Crewe Road, Wythenshawe, Manchester M23 9BE (GB).

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(74) Agents: **HILL, Richard** et al.; Wilson Gunn M'Caw, 41-51 Royal Exchange, Cross Street, Manchester M2 7BD (GB).

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Published:
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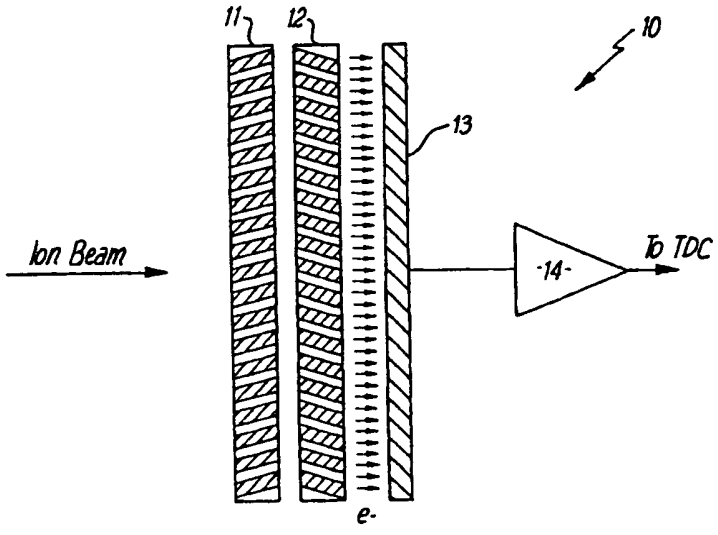
(88) Date of publication of the international search report:
15 November 2001

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

(54) Title: HIGH DYNAMIC RANGE MASS SPECTROMETER



(57) Abstract: A mass spectrometer comprises an ion source which produces an ion beam from a substance to be analysed and a detector to detect a quantity of ions incident thereon. The detector includes two elements (16, 18) each of which detect a part of the quantity of ions and an attenuation device attenuates the quantity of ions reaching one of the detector elements. At least one of the detector elements (16, 18) is connected to a time to digital converter (TDC) to allow counting of the ions and at least one of the detector elements is connected in parallel to both a time to digital converter (TDC) and an analogue to digital converter (ADC).

WO 01/18846 A3

INTERNATIONAL SEARCH REPORT

Internal Application No
PCT/GB 00/03332

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IPC 7 H01J49/40 H01J49/02 G01T1/28

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 H01J G01T

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
EPO-Internal, WPI Data, PAJ, INSPEC

C. DOCUMENTS CONSIDERED TO BE RELEVANT

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A	---	2,7,9
A	WO 98 21742 A (DAVIS LARRY J ;ROCKWOOD ALAN (US); SENSAR CORP (US)) 22 May 1998 (1998-05-22) cited in the application abstract	1

☐ Further documents are listed in the continuation of box C.

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* Special categories of cited documents :

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"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
25 July 2001	01/08/2001

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Hulne, S

1

Form PCT/ISA/210 (second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

Information on patent family members

Interna . al Application No
PCT/GB 00/03332

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 9938190 A	29-07-1999	EP 0970504 A	12-01-2000
		EP 0970505 A	12-01-2000
		EP 0970506 A	12-01-2000
		WO 9938191 A	29-07-1999
		WO 9938192 A	29-07-1999
		JP 2001503196 T	06-03-2001
		JP 2000513494 T	10-10-2000
		US 6229142 B	08-05-2001
WO 9821742 A	22-05-1998	US 5777326 A	07-07-1998
		US 6163032 A	19-12-2000
		AU 7181898 A	03-06-1998
		EP 0939970 A	08-09-1999

P. ENT COOPERATION TREA

From the INTERNATIONAL BUREAU

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

To:

Commissioner
US Department of Commerce
United States Patent and Trademark
Office, PCT
2011 South Clark Place Room
CP2/5C24
Arlington, VA 22202
ETATS-UNIS D'AMERIQUE

in its capacity as elected Office

Date of mailing (day/month/year) 21 May 2001 (21.05.01)	
International application No. PCT/GB00/03332	**Applicant's or agent's file reference** RH/P/15339WO
International filing date (day/month/year) 31 August 2000 (31.08.00)	**Priority date** (day/month/year) 03 September 1999 (03.09.99)
Applicant DAVIS, Stephen et al	

1. The designated Office is hereby notified of its election made:

[X] in the demand filed with the International Preliminary Examining Authority on:

03 April 2001 (03.04.01)

[ ] in a notice effecting later election filed with the International Bureau on:

2. The election [X] was

[ ] was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO **34, chemin des Colombettes** **1211 Geneva 20, Switzerland**	Authorized officer Olivia TEFY
Facsimile No.: (41-22) 740.14.35	Telephone No.: (41-22) 338.83.38

GB0003332

Form PCT/IB/331 (July 1992)

does not affect the intensity of the beam to a sufficient extent to significantly enhance dynamic range without a very large number of TDC's.

It is an object of the present invention to provide an alternative form of mass spectrometer in which ion counting can be used to cover a wide dynamic range using a small number of TDC's.

Thus and in accordance with the present invention therefore there is provided a mass spectrometer comprising an ion source to produce ions from a substance to be detected and detector means to detect a quantity of ions incident on said detections means wherein the said detection means includes at least two detector elements, each of which elements detect at least a part of said quantity of ions from the ion source and attenuation means which acts to attenuate the quantity of ions reaching at least one said detection element.

With this arrangement it is possible to measure the quantity of ions with and without attenuation which means that both single and multiple ion detections can be quantified more accurately and a high dynamic range for the mass spectrometer can be achieved. This is achieved by parallel acquisition or interleaved acquisition of signal from ion beams with significant attenuation at one detector element and almost no attenuation at another.

Preferably each detector element comprises a separate plate anode. Each detector element may be connected via an amplifier to a time to digital

converter (TDC) to allow counting of detected ions.

Although the discussion has been in terms of using TDC acquisition it will be appreciated that the same principle of attenuation of signal to other detector elements could also be applied to extension of dynamic range using analogue-to-digital conversion (ADC) or combinations of TDC and ADC.

The detector elements may be disposed one behind the other relative to the ion source or alternatively may be disposed one above the other in a plane extending generally perpendicular to the direction of ion travel. In the case where the detector element is disposed one behind the other, an earthed member preferably a wire or grid may be provided between the elements to minimise capacitative coupling between these elements.

The attenuation means may be performed by at least one of the detector elements and in this case the at least one detector element is adapted to allow a proportion of incident signal to pass through the element without being detected. The adaptation may comprise a plurality of perforations or other apertures in the element. Alternatively a separate attenuation device may be provided between the ion source and the detector elements which acts to reduce the number of ions reaching at least one of said elements or at least a part thereof. In these circumstances the attenuation device may comprise a perforated plate.

Preferably, in the case where the attenuation means is formed by a perforation of the detector element, the cross-sectional area of the

perforations compared to the total cross-sectional area of the plate is approximately 1 to 100.

The invention will now be described further by way of example and with reference to the accompany drawings of which:-

Fig. 1 shows a schematic version of a prior art form of mass spectrometer;

Fig. 2 shows a schematic version of one embodiment of mass spectrometer in accordance with the present invention;

Fig. 3 shows a variation on the embodiment shown in Fig. 2;

Fig. 4 shows a schematic version of a second embodiment of mass spectrometer in accordance with the present invention;

Fig. 5 shows a schematic version of a third embodiment of mass spectrometer in accordance with the present invention;

Fig. 6 shows a schematic version of a fourth embodiment of mass spectrometer in accordance with the present invention; and

Fig. 7 shows a schematic version of a fifth embodiment of mass spectrometer in accordance with the present invention.

Referring now to the drawings, there is shown in Fig. 1 a schematic representation of one standard form of prior art mass spectrometer detector. The spectrometer 10 comprises an ion source (not shown) which produces an ion beam from a substance to be analysed. The ion beam is directed by conventional means onto a pair of microchannel plates 11,12 (hereinafter

A further alternative is shown in Fig. 5. In this embodiment, the first anode 16, a second anode 18 and, optionally an earthed grid 19, are constructed as sandwich layers of a printed circuit board 21. The first anode 16 is formed as a perforated plate attached to a first support layer 22 which is also perforated, the perforations in the first support layer 22 being in register with the perforations in the first anode 16. Attached to the opposite side of the first support layer 22 is an earthed gird, perforations in the grid also being in register with the perforations in the first support layer 22 and the first anode 16. Attached to the opposite side of the earthed grid 19 is a second support layer 23 which carries a second anode 18 attached thereto. Fingers 24 of the second anode 18 extend through the second support layer 23 and terminate adjacent to the perforations in the earthed grid 19.

In this embodiment, the attenuation is carried out by the first anode 16 and only a proportion of the secondary electrons reach the fingers 24 of the second anode 18 through the aligned apertures. As in the previous embodiments, the earthed grid 19 minimises capacitative coupling between the two anodes.

A still further alternative is shown in Fig. 6 in which a separate attenuation element 26 of appropriate form is placed in the ion beam before the ion beam is incident on the chevron pair 11,12. The attenuation element in this embodiment, comprises a perforated plate, and is arranged

CLAIMS

1. A mass spectrometer comprising an ion source to produce ions from a substance to be detected and detector means to detect a quantity of ions incident on said detection means wherein the said detection means includes at least two detector elements, each of which elements detect at least a part of said quantity of ions from the ion source and attenuation means which acts to attenuate the quantity of ions reaching at least one said detection element, wherein at least one of said detection elements is connected to a time-to-digital converter (TDC) to allow counting of detected ions and at least one of said detection elements is connected in parallel to both a time-to-digital converter (TDC) and an analogue-to-digital converter (ADC) for ion detection.

2. A mass spectrometer according to Claim 1, wherein attenuation means is such that both incident ions and secondary electrons generated by said incident ions are attenuated.

3. A mass spectrometer according to Claim 1 or Claim 2, wherein each detector element comprises a separate plate anode.

4. A mass spectrometer according to any one of Claims 1 to 3, wherein each detector element is connected via an amplifier to a time to digital converter (TDC) to allow counting of detected ions.

5. A mass spectrometer according to any one of Claims 1 to 4, wherein

the detector elements are disposed one behind the other relative to the ion source.

6. A mass spectrometer according to any one of Claims 1 to 4, wherein the detector elements are disposed one above the other in a plane extending generally perpendicular to the direction of ion travel.

7. A mass spectrometer according to Claim 5, wherein an earthed grid is provided between the elements to minimise capacitative coupling between elements.

8. A mass spectrometer according to any one of Claims 1 to 7, wherein the attenuation means is formed by at least one of the detector elements.

9. A mass spectrometer according to Claim 8, wherein the at least one detector element is adapted to allow a proportion of incident signal to pass through the element without being detected.

10. A mass spectrometer according to Claim 9, wherein the adaptation of the at least one detector comprises a plurality of perforations or other apertures in the element.

11. A mass spectrometer according to any one of claims 1 to 8, wherein said attenuation device is provided between the ion source and the detector elements which acts to reduce the number of ions reaching at least one of said elements or at least a part thereof.

12. A mass spectrometer according to Claim 11, wherein the attenuation

device comprises a perforated plate.

13. A mass according to claim 10, wherein the cross-sectional area of the perforations compared to the total cross-sectional area of the plate is approximately 1 to 100.

14. A mass spectrometer substantially as hereinbefore described with reference to the accompanying drawings.

does not affect the intensity of the beam to a sufficient extent to significantly enhance dynamic range without a very large number of TDC's.

It is an object of the present invention to provide an alternative form of mass spectrometer in which ion counting can be used to cover a wide dynamic range using a small number of TDC's.

Thus and in accordance with the present invention therefore there is provided a mass spectrometer comprising a mass spectrometer comprising an ion source to produce ions from a substance to be detected and detector means to detect a quantity of ions incident on said detection means wherein the said detection means includes at least two detector elements, each of which elements detect at least a part of said quantity of ions from the ion source and attenuation means which acts to attenuate the quantity of ions reaching at least one said detection element, wherein at least one of said detection elements is connected to a time-to-digital converter (TDC) to allow counting of detected ions and at least one of said detection elements is connected in parallel to both a time-to-digital converter (TDC) and an analogue-to-digital converter (ADC) for ion detection.

With this arrangement it is possible to measure the quantity of ions with and without attenuation which means that both single and multiple ion detections can be quantified more accurately and a high dynamic range for the mass spectrometer can be achieved. This is achieved by parallel acquisition or interleaved acquisition of signal from ion beams with

significant attenuation at one detector element and almost no attenuation at another.

Although the discussion has been in terms of using TDC acquisition it will be appreciated that the same principle of attenuation of signal to other detector elements could also be applied to extension of dynamic range using analogue-to-digital conversion (ADC) or combinations of TDC and ADC.

The detector elements may be disposed one behind the other relative to the ion source or alternatively may be disposed one above the other in a plane extending generally perpendicular to the direction of ion travel. In the case where the detector element is disposed one behind the other, an earthed member preferably a wire or grid may be provided between the elements to minimise capacitative coupling between these elements.

The attenuation means may be performed by at least one of the detector elements and in this case the at least one detector element is adapted to allow a proportion of incident signal to pass through the element without being detected. The adaptation may comprise a plurality of perforations or other apertures in the element. Alternatively a separate attenuation device may be provided between the ion source and the detector elements which acts to reduce the number of ions reaching at least one of said elements or at least a part thereof. In these circumstances the attenuation device may comprise a perforated plate.

Preferably, in the case where the attenuation means is formed by a

perforation of the detector element, the cross-sectional area of the perforations compared to the total cross-sectional area of the plate is substantially 1 to 100.

The invention will now be described further by way of example and with reference to the accompany drawings of which:-

Fig. 1 shows a schematic version of a prior art form of mass spectrometer;

Fig. 2 shows a schematic version of one embodiment of mass spectrometer;

Fig. 3 shows a variation on the embodiment shown in Fig. 2;

Fig. 4 shows a schematic version of a second embodiment of mass spectrometer;

Fig. 5 shows a schematic version of a third embodiment of mass spectrometer;

Fig. 6 shows a schematic version of a fourth embodiment of mass spectrometer in accordance with the present invention; and

Fig. 7 shows a schematic version of a fifth embodiment of mass spectrometer in accordance with the present invention.

Referring now to the drawings, there is shown in Fig. 1 a schematic representation of one standard form of prior art mass spectrometer detector. The spectrometer 10 comprises an ion source (not shown) which produces an ion beam from a substance to be analysed. The ion beam is directed by conventional means onto a pair of microchannel plates 11,12 (hereinafter

A further alternative is shown in Fig. 5. In this embodiment, the first anode 16, a second anode 18 and, optionally an earthed grid 19, are constructed as sandwich layers of a printed circuit board 21. The first anode 16 is formed as a perforated plate attached to a first support layer 22 which is also perforated, the perforations in the first support layer 22 being in register with the perforations in the first anode 16. Attached to the opposite side of the first support layer 22 is an earthed gird, perforations in the grid also being in register with the perforations in the first support layer 22 and the first anode 16. Attached to the opposite side of the earthed grid 19 is a second support layer 23 which carries a second anode 18 attached thereto. Fingers 24 of the second anode 18 extend through the second support layer 23 and terminate adjacent to the perforations in the earthed grid 19.

In this embodiment, the attenuation is carried out by the first anode 16 and only a proportion of the secondary electrons reach the fingers 24 of the second anode 18 through the aligned apertures. As in the previous embodiments, the earthed grid 19 minimises capacitative coupling between the two anodes.

The embodiments of Figs. 2-5 are not embodiments of mass spectrometer in accordance with the present invention.

A still further alternative is shown in Fig. 6 in which a separate attenuation element 26 of appropriate form is placed in the ion beam before the ion beam is incident on the chevron pair 11,12. The attenuation element in this embodiment, comprises a perforated plate, and is arranged

CLAIMS

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2. A mass spectrometer according to Claim 1, wherein said attenuation means also acts to attenuate secondary electrons generated by said ions incident on said detection means.

3. A mass spectrometer according to Claim 1 or Claim 2, wherein each detector element comprises a separate plate anode.

4. A mass spectrometer according to any one of Claims 1 to 3, wherein each detector element is connected via an amplifier to a time to digital converter (TDC) to allow counting of detected ions.

5. A mass spectrometer according to any one of Claims 1 to 4, wherein

the detector elements are disposed one behind the other relative to the ion source.

6. A mass spectrometer according to any one of Claims 1 to 4, wherein the detector elements are disposed one above the other in a plane extending generally perpendicular to the direction of ion travel.

7. A mass spectrometer according to Claim 5, wherein an earthed grid is provided between the elements to minimise capacitative coupling between elements.

8. A mass spectrometer according to any one of Claims 1 to 7, wherein the attenuation means is formed by at least one of the detector elements.

9. A mass spectrometer according to Claim 8, wherein the adaptation of the at least one detector comprises a plurality of perforations or other apertures in the element to allow a proportion of incident signal to pass through the element without being detected.

10. A mass spectrometer according to any one of claims 1 to 9, wherein said attenuation device is provided between the ion source and the detector elements which acts to reduce the number of ions reaching at least one of said elements or at least a part thereof.

11. A mass spectrometer according to Claim 10, wherein the attenuation device comprises a perforated plate.

12. A mass spectrometer according to claim 9, wherein the cross-

sectional area of the perforations compared to the total cross-sectional area of the plate is substantially 1 to 100.

CLAIMS

1. A mass spectrometer comprising an ion source to produce ions from a substance to be detected and detector means to detect a quantity of ions incident on said detection means wherein the said detection means includes at least two detector elements, each of which elements detect at least a part of said quantity of ions from the ion source and attenuation means which acts to attenuate the quantity of ions reaching at least one said detection element, wherein at least one of said detection elements is connected to a time-to-digital converter (TDC) to allow counting of detected ions and at least one of said detection elements is connected in parallel to both a time-to-digital converter (TDC) and an analogue-to-digital converter (ADC) for ion detection characterised in that said attenuation means acts to allow only those ions to be detected that produce secondary electrons that are incident only on one or other of said detection elements.

2. A mass spectrometer according to Claim 1, wherein said attenuation means also acts to attenuate secondary electrons generated by said ions incident on said detection means.

3. A mass spectrometer according to Claim 1 or Claim 2, wherein each detector element comprises a separate plate anode.

4. A mass spectrometer according to any one of Claims 1 to 3, wherein each detector element is connected via an amplifier to a time to

digital converter (TDC) to allow counting of detected ions.

5. A mass spectrometer according to any one of Claims 1 to 4, wherein the detector elements are disposed one behind the other relative to the ion source.

6. A mass spectrometer according to any one of Claims 1 to 4, wherein the detector elements are disposed one above the other in a plane extending generally perpendicular to the direction of ion travel.

7. A mass spectrometer according to Claim 5, wherein an earthed grid is provided between the elements to minimise capacitative coupling between elements.

8. A mass spectrometer according to any one of Claims 1 to 7, wherein the attenuation means is formed by at least one of the detector elements.

9. A mass spectrometer according to Claim 8, wherein the adaptation of the at least one detector comprises a plurality of perforations or other apertures in the element to allow a proportion of incident signal to pass through the element without being detected.

10. A mass spectrometer according to any one of claims 1 to 9, wherein said attenuation device is provided between the ion source and the detector elements which acts to reduce the number of ions reaching at least one of said elements or at least a part thereof.

11. A mass spectrometer according to Claim 10, wherein the attenuation device comprises a perforated plate.

12. A mass spectrometer according to claim 9, wherein the cross-sectional area of the perforations compared to the total cross-sectional area of the plate is substantially 1 to 100.

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(54) Title: HIGH DYNAMIC RANGE MASS SPECTROMETER

(57) Abstract: A mass spectrometer comprises an ion source which produces an ion beam from a substance to be analysed and a detector to detect a quantity of ions incident thereon. The detector includes two elements (16, 18) each of which detect a part of the quantity of ions and an attenuation device attenuates the quantity of ions reaching one of the detector elements. At least one of the detector elements (16, 18) is connected to a time to digital converter (TDC) to allow counting of the ions and at least one of the detector elements is connected in parallel to both a time to digital converter (TDC) and an analogue to digital converter (ADC).

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HIGH DYNAMIC RANGE MASS SPECTROMETER

This invention relates to a high dynamic range mass spectrometer, preferably although not exclusively of the time of flight kind.

Time of flight (TOF) mass spectrometers are often used for quantitative analysis of substances. In these applications of a TOF mass spectrometer, it will be necessary to be able to accurately determine the concentration of a substance based upon a detected ion signal. In a TOF mass spectrometer, the ion signals which are to be detected are usually fast transients and can be measured by analogue to digital conversion using a transient recorder or by ion counting as a function of time using a time to digital convertor (TDC). Use of a TDC is generally preferred because it can be more difficult to obtain accurate quantitative results using a transient recorder. The use of ion counting is further preferred in an orthogonal acceleration TOF because the signals to be measured tend to be small and the ion count rates are low. Ion counting using a TDC involves the TDC detecting the presence of a signal at the detector in excess of a predetermined threshold. If the signal detected is in excess of a predetermined threshold then this is deemed to be indicative of the presence of an ion at the detector and the TDC, after detection of the above threshold signal, increments a counter to count the ions.

However, a problem arises with a time to digital convertor when this is used to count ions in intense ion beams because most TDC's can only

detect one event in a finite small time window. This means that where a TDC is used, it is not normally possible to distinguish between a single ion being detected and a multiplicity of ions being detected at the same time. This arises because a TDC cannot distinguish between different magnitudes of signal, only whether the detected signal exceeds the predetermined threshold. Accordingly, a counter connected to the TDC will only be incremented once upon detection of an above threshold signal regardless of its magnitude and therefore in the case of intense ion beams an accurate quantitative measurement cannot be made. This means that mass spectrometers incorporating such ion counters usually require there to be less than or equal to one ion per signal pulse of any substance to measured. It also means that for a single TDC there will be a relatively low dynamic range.

Attempts have been made to provide a mass spectrometer which uses one or more TDC's to count ions and in which the dynamic range can be extended for better quantitative measurements.

Thus for example, U.S. Patent No. 5,777,326 discloses a TOF mass spectrometer in which the incoming ion beam is spread so as to be capable of being detected by three or more detectors. The signal at each detector is detected by a respective TDC and the signal from each TDC is subsequently added together. However, the problem with this type of arrangement is that simply spreading the beam over a number of detectors

does not affect the intensity of the beam to a sufficient extent to significantly enhance dynamic range without a very large number of TDC's.

It is an object of the present invention to provide an alternative form of mass spectrometer in which ion counting can be used to cover a wide dynamic range using a small number of TDC's.

Thus and in accordance with the present invention therefore there is provided a mass spectrometer comprising an ion source to produce ions from a substance to be detected and detector means to detect a quantity of ions incident on said detections means wherein the said detection means includes at least two detector elements, each of which elements detect at least a part of said quantity of ions from the ion source and attenuation means which acts to attenuate the quantity of ions reaching at least one said detection element.

With this arrangement it is possible to measure the quantity of ions with and without attenuation which means that both single and multiple ion detections can be quantified more accurately and a high dynamic range for the mass spectrometer can be achieved. This is achieved by parallel acquisition or interleaved acquisition of signal from ion beams with significant attenuation at one detector element and almost no attenuation at another.

Preferably each detector element comprises a separate plate anode. Each detector element may be connected via an amplifier to a time to digital

converter (TDC) to allow counting of detected ions.

Although the discussion has been in terms of using TDC acquisition it will be appreciated that the same principle of attenuation of signal to other detector elements could also be applied to extension of dynamic range using analogue-to-digital conversion (ADC) or combinations of TDC and ADC.

The detector elements may be disposed one behind the other relative to the ion source or alternatively may be disposed one above the other in a plane extending generally perpendicular to the direction of ion travel. In the case where the detector element is disposed one behind the other, an earthed member preferably a wire or grid may be provided between the elements to minimise capacitative coupling between these elements.

The attenuation means may be performed by at least one of the detector elements and in this case the at least one detector element is adapted to allow a proportion of incident signal to pass through the element without being detected. The adaptation may comprise a plurality of perforations or other apertures in the element. Alternatively a separate attenuation device may be provided between the ion source and the detector elements which acts to reduce the number of ions reaching at least one of said elements or at least a part thereof. In these circumstances the attenuation device may comprise a perforated plate.

Preferably, in the case where the attenuation means is formed by a perforation of the detector element, the cross-sectional area of the

perforations compared to the total cross-sectional area of the plate is approximately 1 to 100.

The invention will now be described further by way of example and with reference to the accompany drawings of which:-

Fig. 1 shows a schematic version of a prior art form of mass spectrometer;

Fig. 2 shows a schematic version of one embodiment of mass spectrometer in accordance with the present invention;

Fig. 3 shows a variation on the embodiment shown in Fig. 2;

Fig. 4 shows a schematic version of a second embodiment of mass spectrometer in accordance with the present invention;

Fig. 5 shows a schematic version of a third embodiment of mass spectrometer in accordance with the present invention;

Fig. 6 shows a schematic version of a fourth embodiment of mass spectrometer in accordance with the present invention; and

Fig. 7 shows a schematic version of a fifth embodiment of mass spectrometer in accordance with the present invention.

Referring now to the drawings, there is shown in Fig. 1 a schematic representation of one standard form of prior art mass spectrometer detector. The spectrometer 10 comprises an ion source (not shown) which produces an ion beam from a substance to be analysed. The ion beam is directed by conventional means onto a pair of microchannel plates 11,12 (hereinafter

referred to as a chevron pair) which generates secondary electrons due to the collision of the ions in the ion beam with the material of the plates 11,12 in the microchannels. Secondary electrons generated are detected by a single plate anode 13, the detected signal is amplified in an amplifier 14 and is passed to a time to digital convertor (TDC) (not shown) which detects detected signals over a predetermined threshold and increments a counter to count these above threshold signals.

This form of mass spectrometer suffers from the problem that if an above threshold signal is detected by the TDC, the counter will be incremented only once regardless of the magnitude of the signal in exceeding the threshold. Thus even if the signal is of such a magnitude as to constitute more than one ion being detected, the counter will still only be incremented once. The TDC cannot distinguish between different magnitude above threshold signals. This means that the mass spectrometer is very inaccurate when used for quantitative measurements of intense signals.

One form of mass spectrometer in accordance with the present invention is shown in schematic form in Fig. 2. In this arrangement, the ion beam generated by the ion source (not shown) is also incident on a chevron pair 11,12 as with the embodiment of Fig. 1. The ion beam strikes the microchannel plate 11 and causes the ejection of secondary electrons from the surface of the microchannels. The secondary electrons cause the

ejection of further secondary electrons as they accelerate through the microchannels in the plates 11,12 which results in an electron beam which emerges from the chevron pair 11,12 being essentially an amplified signal version of the incoming ion beam. The secondary electron beam then strikes a first anode 16 for detection. The first anode 16 is perforated in order that some of the secondary electrons pass through the first anode 16 without being detected. The remainder of the secondary electrons strike the first anode 16 and are detected. For detection purposes, the first anode 16 is connected to an amplifier 14 and to a time to digital converter (not shown) the output of which increments a counter (not shown) as previously explained. Those secondary electrons which pass through the perforations 17 in the first anode 16 strike a second anode 18 placed substantially immediately behind the first anode 16 and are detected. The secondary anode is connected to a second amplifier and a second time to digital converter, the output of which increments a counter in the same manner as mentioned above.

It will be appreciated that the ratio of the cross-sectional area of the perforations to the total cross-sectional area of the anode can be chosen to give a particular degree of attenuation to the incoming secondary electron beam.

Thus, in use, the ion beam is directed onto the chevron pair 11,12. This results in the generation of secondary electrons in the manner

mentioned above. These secondary electrons emerge from the chevron pair 11,12 and are incident of the first anode 16. It is thought that by arranging for the cross-sectional area of the perforations in the first anode to be of the order of 1% of the total cross-sectional area of the anode will give the possibility for more accurate quantitative measurements over a large dynamic range, however, it is to be appreciated that the ratio of the cross-sectional area of the perforations to the total area of the anode can be of any desired magnitude in order to give appropriate attenuation characteristics.

Therefore, if the area of the perforations represents approximately 1% of the total area of the anode, this means that 1% of the secondary electron beam which is incident on the first anode 16 will pass through that anode without being detected. This means that the intensity of any signal present at the first anode would be reduced by two orders of magnitude if measured at the second anode 18. Therefore it would be appreciated that with this arrangement, that if for example the first anode 16 can be used to detect signals of a first two orders of magnitude then the second anode, at which the signal has been reduced in intensity by a factor of 100, can be used to detect signals at a second two orders of magnitude. It will be appreciated that this allows much more accurate quantitative analysis of the incoming ion beam since signals which are above threshold will be differentiated according to their magnitude and accordingly if a signal is of such a

magnitude as to constitute more than one ion arriving, the present arrangement will detect this and the counters will be incremented by the respective TDC's by the correct number of ions. It can clearly be seen that this will result in a significant increase in the dynamic range of the mass spectrometer.

Fig. 3 shows a variation on the embodiment of Fig. 2 in which an earthed grid 19 is positioned between the first and second anode 16 and 18. The earthed grid 19 assists in the minimisation of capacitative coupling effects between the two anodes 16 and 18.

Whilst in the embodiments of Figs. 2 and 3, attenuation of the secondary electron signal is carried out by the perforated first anode 16, attenuation can be carried out in many different ways.

Thus for example, as shown in Fig. 4, the attenuation can be carried out by wires or a grid placed in front of the first anode 16 to form the second anode 18. The cross-sectional area of the wire or grid compared to the cross-sectional area of the first plate anode is small such that a large proportion of the incident signal from the chevron pair 11,12 passes through the second anode 18 without being detected. As with the other embodiments, the attenuation can be varied by changing the cross-sectional area of the wire or grid to achieve a desired dynamic range. Furthermore, as with the other embodiments, an earthed grid 19 can be placed between the two anodes to minimise capacitative coupling of these anodes.

A further alternative is shown in Fig. 5. In this embodiment, the first anode 16, a second anode 18 and, optionally an earthed grid 19, are constructed as sandwich layers of a printed circuit board 21. The first anode 16 is formed as a perforated plate attached to a first support layer 22 which is also perforated, the perforations in the first support layer 22 being in register with the perforations in the first anode 16. Attached to the opposite side of the first support layer 22 is an earthed gird, perforations in the grid also being in register with the perforations in the first support layer 22 and the first anode 16. Attached to the opposite side of the earthed grid 19 is a second support layer 23 which carries a second anode 18 attached thereto. Fingers 24 of the second anode 18 extend through the second support layer 23 and terminate adjacent to the perforations in the earthed grid 19.

In this embodiment, the attenuation is carried out by the first anode 16 and only a proportion of the secondary electrons reach the fingers 24 of the second anode 18 through the aligned apertures. As in the previous embodiments, the earthed grid 19 minimises capacitative coupling between the two anodes.

A still further alternative is shown in Fig. 6 in which a separate attenuation element 26 of appropriate form is placed in the ion beam before the ion beam is incident on the chevron pair 11,12. The attenuation element in this embodiment, comprises a perforated plate, and is arranged

so as to interfere only with a part of the incoming ion beam and reduces the proportion of that part of the beam which reaches the chevron pair 11,12. In this embodiment, the first anode 16 and the second anode 18 are also provided but they are provided in the same plane extending generally parallel to the longitudinal axis of the chevron pair 11,12 as spaced therefrom. Thus the attenuation element attenuates only a part of the incoming ion beam which, after passing through the chevron pair 11,12 and generating secondary electrons, is incident on the second anode 18. The unattenuated part of the incoming ion beam after passing through the chevron pair 11,12 is incident on the first anode 16. Therefore it will be appreciated that the same effect is achieved with this embodiment as is achieved in the other embodiments.

It will of course be appreciated that the overall attenuation required may also be achieved by a combination of attenuation of the incident ion beam reaching an area of the microchannel plates detector and attenuation of the secondary electron signal, for example Fig. 7.

It will further be appreciated that attenuation can be achieved by a combination of restricting the proportion of ion beam reaching a part of the chevron pair 11,12 (as in the embodiment of Fig. 6) with a restriction on the secondary electron signal emerging from the chevron pair (as in the embodiment of Fig. 4). An example of an embodiment of this type is shown in Fig. 7. In this embodiment, the incident ion beam is attenuated by a

perforated member placed before the chevron pair 11,12. Also the secondary electron signal emerging from the chevron pair 11,12 is attenuated by placing a relatively small second anode in front of an relatively large first anode.

It will be appreciated that it is the attenuation of the incoming ion beam or the secondary electrons ejected from the chevron pair 11,12 which allows the TDC elements to more accurately count incoming ions over a large dynamic range. The use of attenuation means that it is possible to discriminate between different magnitude above threshold signals giving rise to a more accurate quantitative analysis of the incoming ion beam and also giving rise to an extension to the dynamic range of the mass spectrometer.

It is of course to be understood that the invention is not intended to be restricted to the details of the above embodiment which are described by way of example only.

CLAIMS

1. A mass spectrometer comprising an ion source to produce ions from a substance to be detected and detector means to detect a quantity of ions incident on said detection means wherein the said detection means includes at least two detector elements, each of which elements detect at least a part of said quantity of ions from the ion source and attenuation means which acts to attenuate the quantity of ions reaching at least one said detection element, wherein at least one of said detection elements is connected to a time-to-digital converter (TDC) to allow counting of detected ions and at least one of said detection elements is connected in parallel to both a time-to-digital converter (TDC) and an analogue-to-digital converter (ADC) for ion detection.

2. A mass spectrometer according to Claim 1, wherein attenuation means is such that both incident ions and secondary electrons generated by said incident ions are attenuated.

3. A mass spectrometer according to Claim 1 or Claim 2, wherein each detector element comprises a separate plate anode.

4. A mass spectrometer according to any one of Claims 1 to 3, wherein each detector element is connected via an amplifier to a time to digital converter (TDC) to allow counting of detected ions.

5. A mass spectrometer according to any one of Claims 1 to 4, wherein

the detector elements are disposed one behind the other relative to the ion source.

6. A mass spectrometer according to any one of Claims 1 to 4, wherein the detector elements are disposed one above the other in a plane extending generally perpendicular to the direction of ion travel.

7. A mass spectrometer according to Claim 5, wherein an earthed grid is provided between the elements to minimise capacitative coupling between elements.

8. A mass spectrometer according to any one of Claims 1 to 7, wherein the attenuation means is formed by at least one of the detector elements.

9. A mass spectrometer according to Claim 8, wherein the at least one detector element is adapted to allow a proportion of incident signal to pass through the element without being detected.

10. A mass spectrometer according to Claim 9, wherein the adaptation of the at least one detector comprises a plurality of perforations or other apertures in the element.

11. A mass spectrometer according to any one of claims 1 to 8, wherein said attenuation device is provided between the ion source and the detector elements which acts to reduce the number of ions reaching at least one of said elements or at least a part thereof.

12. A mass spectrometer according to Claim 11, wherein the attenuation

device comprises a perforated plate.

13. A mass according to claim 10, wherein the cross-sectional area of the perforations compared to the total cross-sectional area of the plate is approximately 1 to 100.

14. A mass spectrometer substantially as hereinbefore described with reference to the accompanying drawings.

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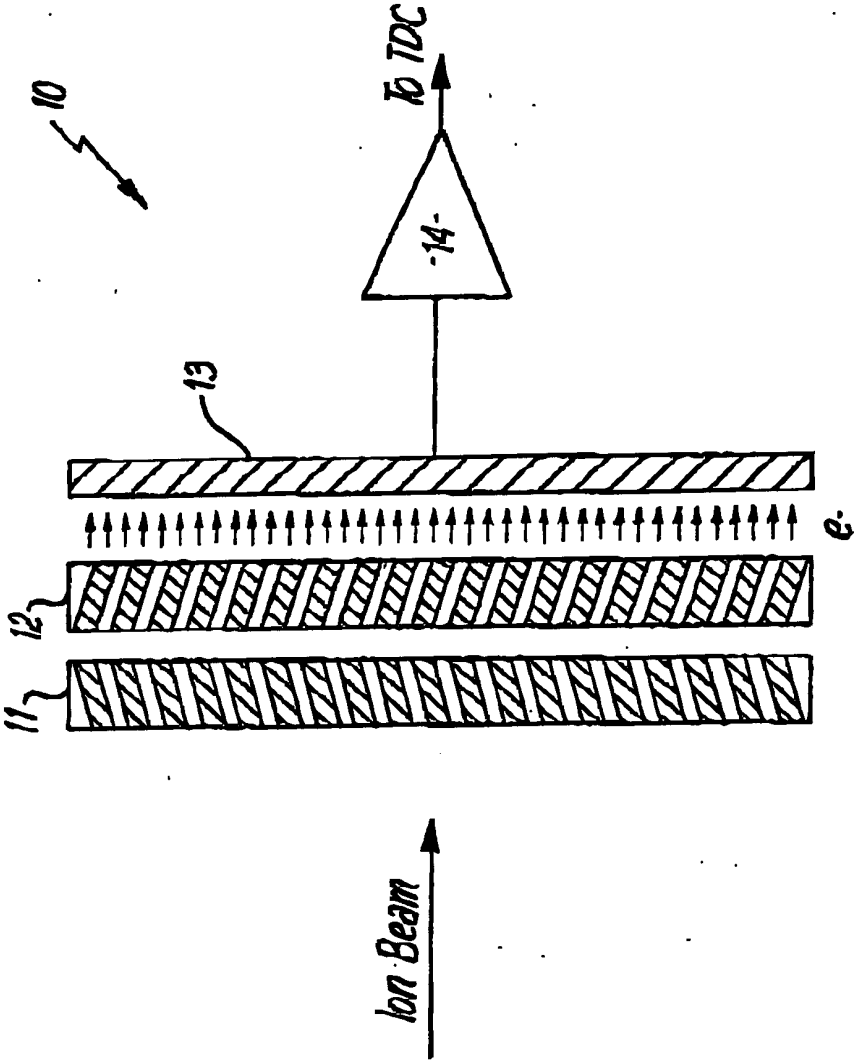

Fig. 1

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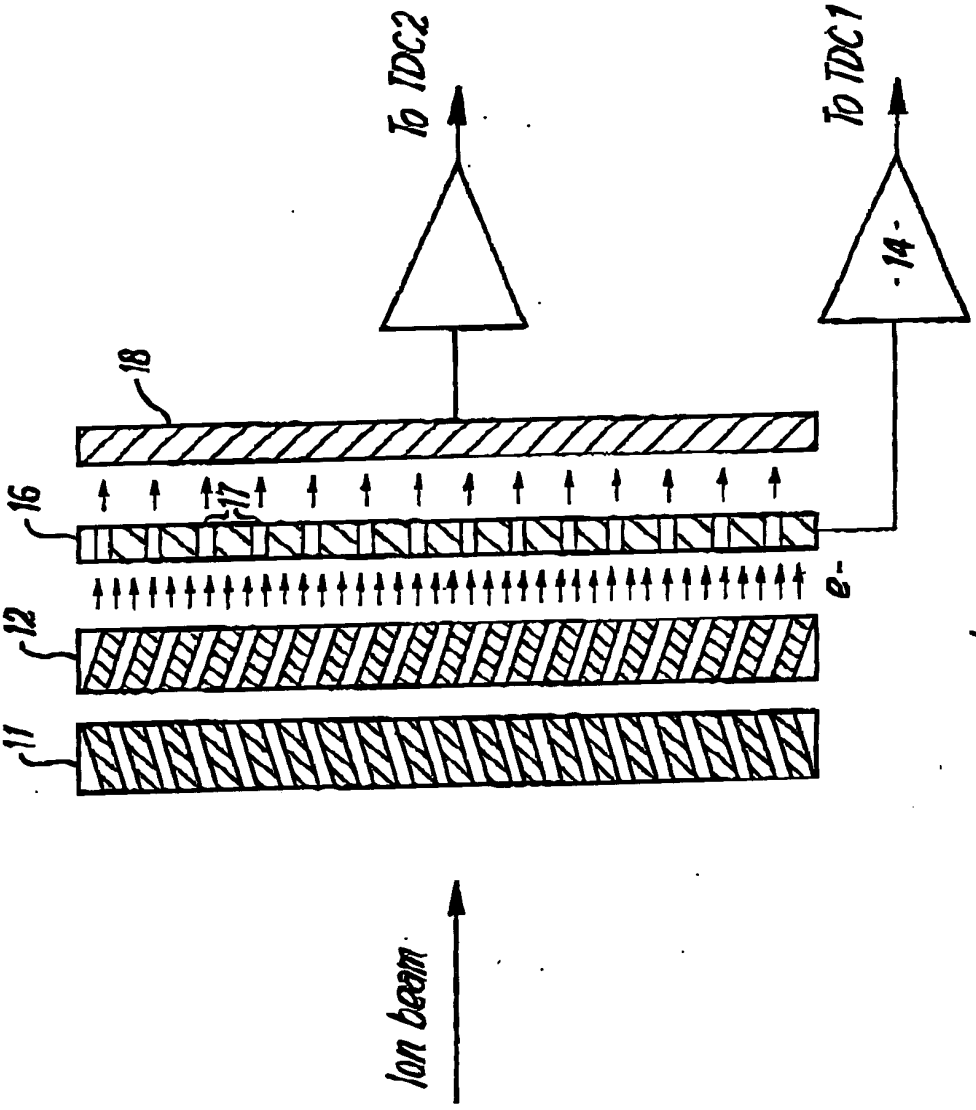

Fig.2

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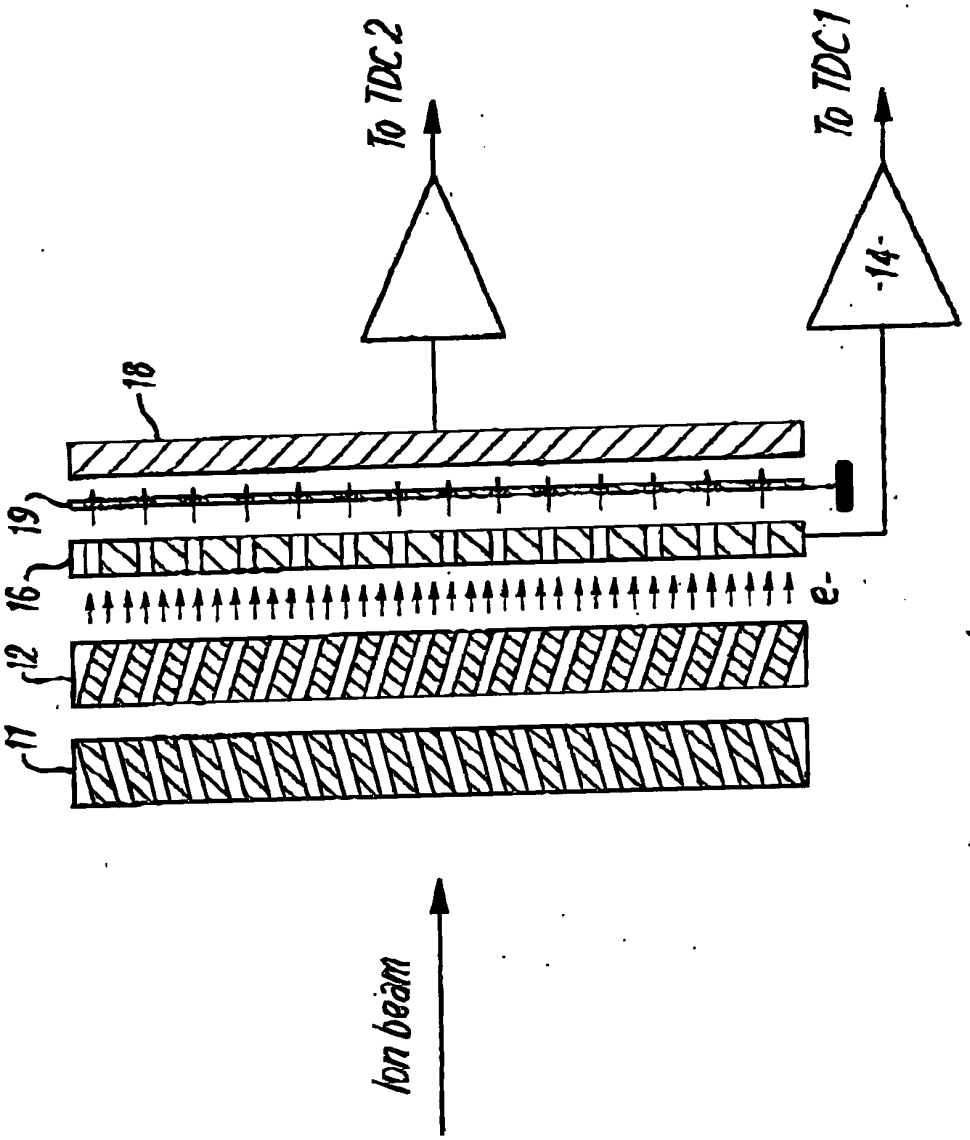

Fig. 3

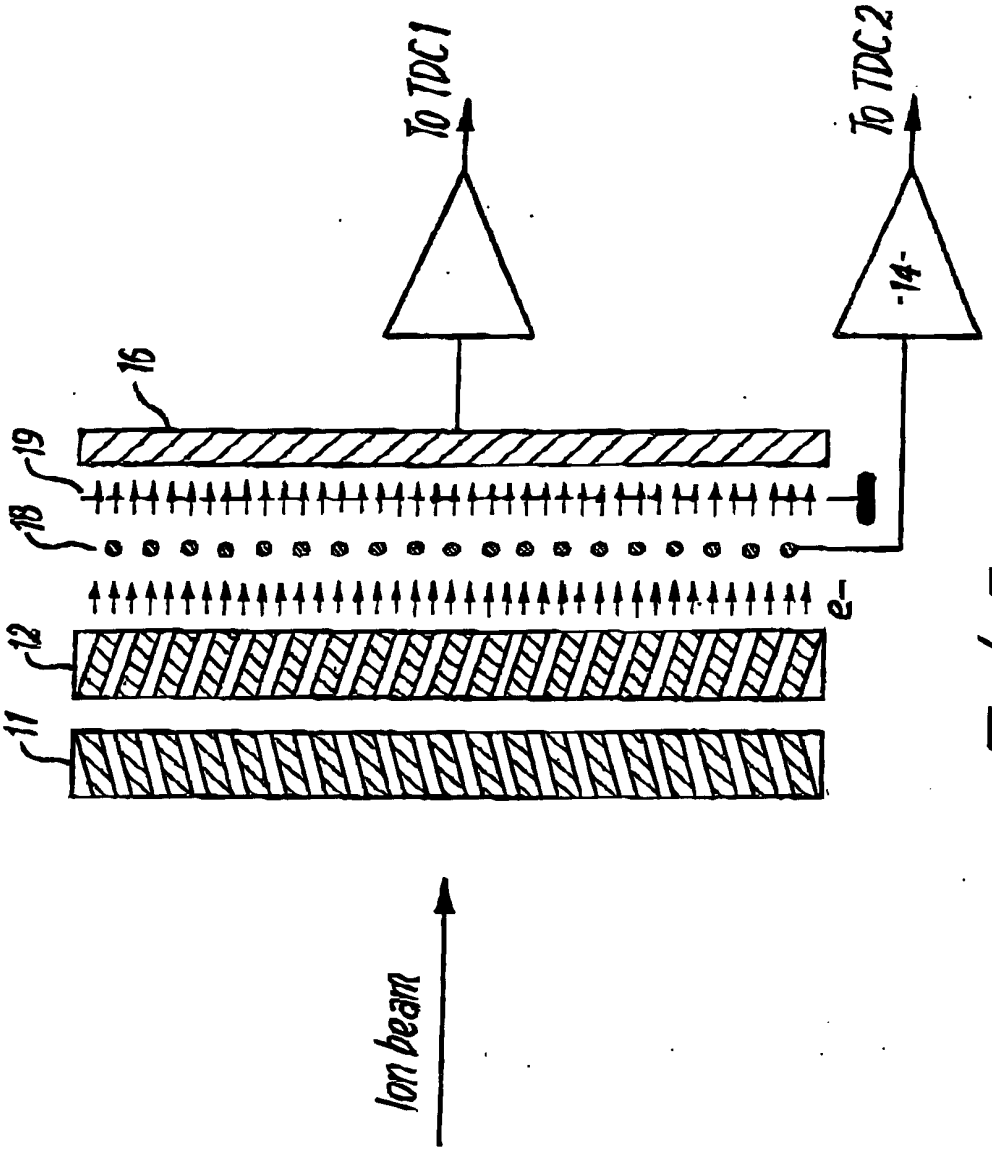

Fig.4

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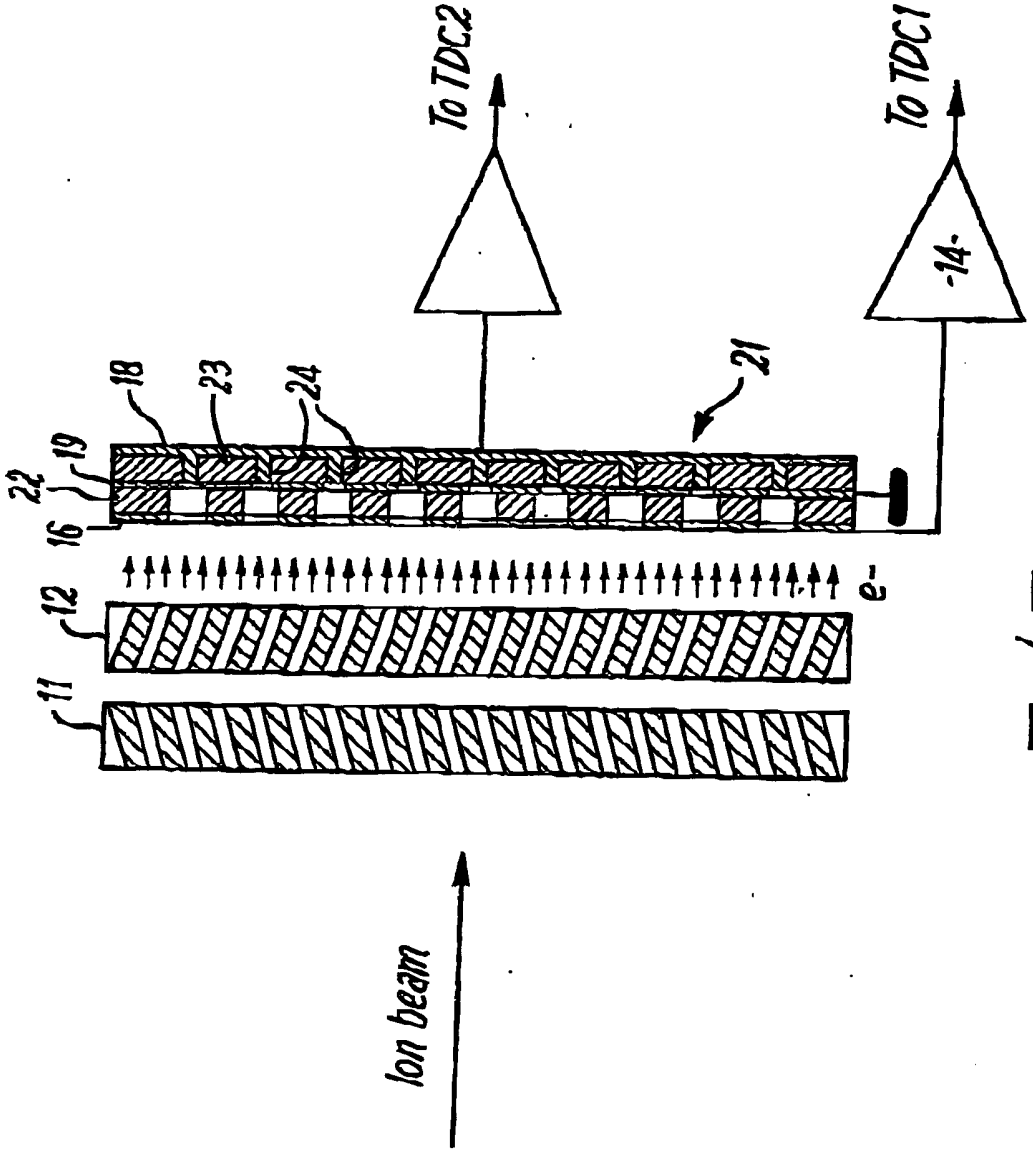

FIG. 5

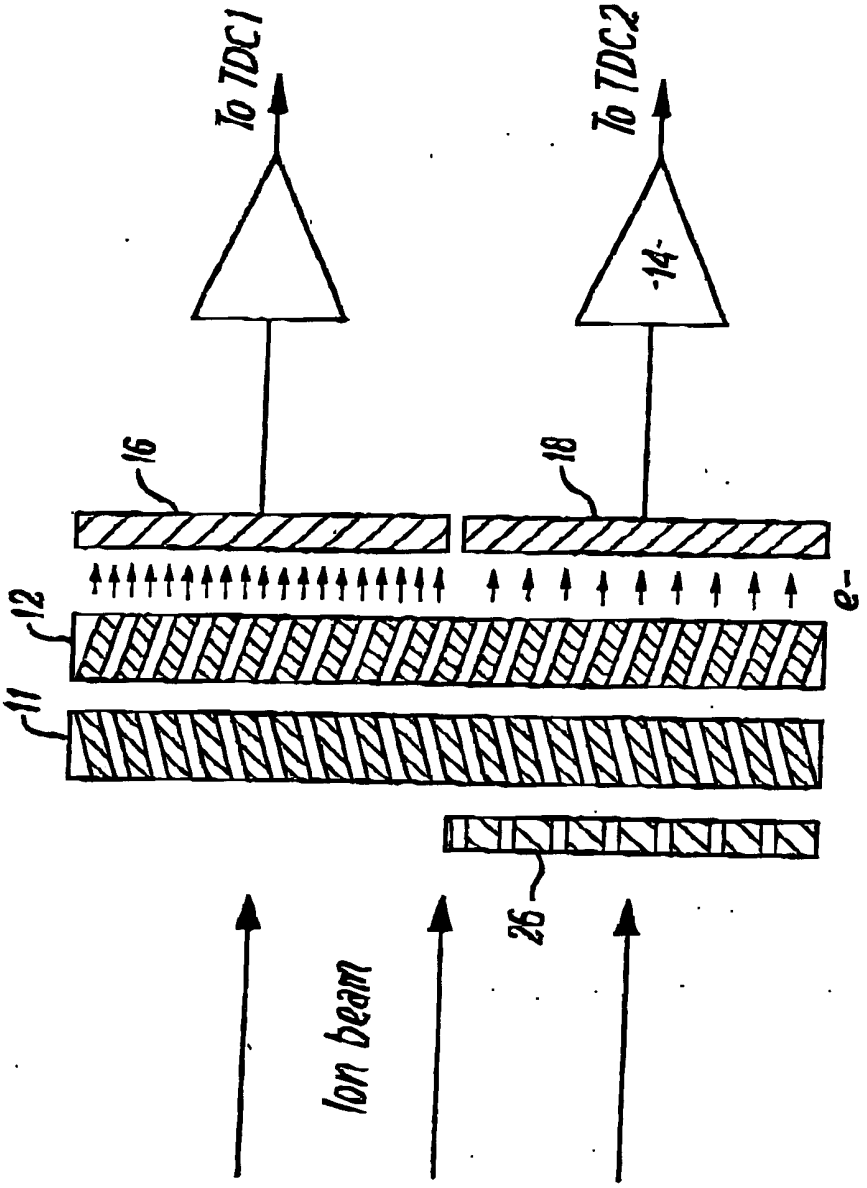

FIG. 6

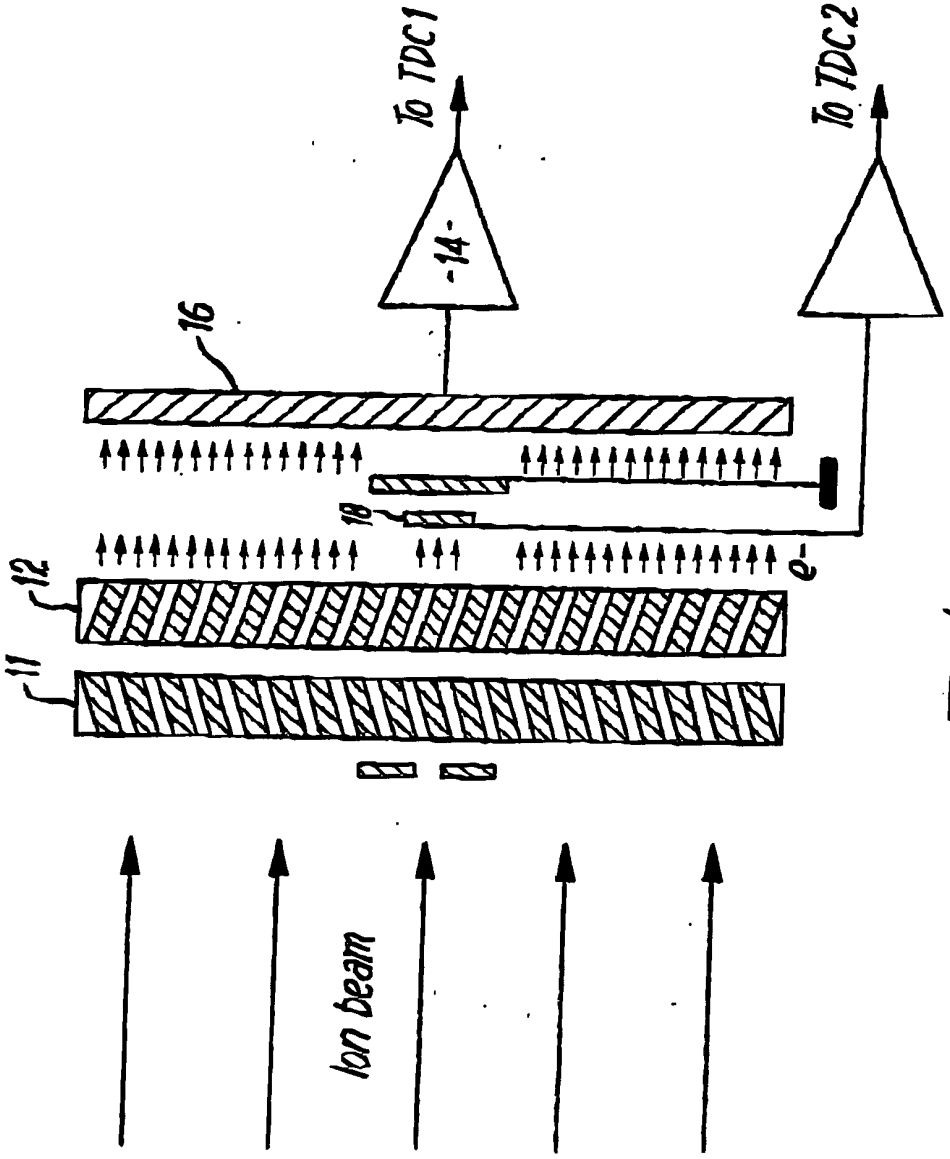

FIG. 1

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PATENT COOPERATION TREATY

PCT

From the INTERNATIONAL SEARCHING AUTHORITY

To:
BOULT WADE TENNANT
Attn. Frost, Alex John
Verulam Gardens
70 Gray's Inn Road
London WC1X 8BT
UNITED KINGDOM

NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL SEARCH REPORT OR THE DECLARATION

(PCT Rule 44.1)

Applicant's or agent's file reference AJF57571/001	Date of mailing *(day/month/year)* 17/12/2002 **FOR FURTHER ACTION** See paragraphs 1 and 4 below
International application No. PCT/GB 02/ 02488	International filing date *(day/month/year)* 28/05/2002
Applicant THERMO FINNIGAN LLC	

1. [X] The applicant is hereby notified that the International Search Report has been established and is transmitted herewith.

Filing of amendments and statement under Article 19:
The applicant is entitled, if he so wishes, to amend the claims of the International Application (see Rule 46):

When? The time limit for filing such amendments is normally 2 months from the date of transmittal of the International Search Report; however, for more details, see the notes on the accompanying sheet.

Where? Directly to the International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland
Fascimile No.: (41-22) 740.14.35

For more detailed instructions, see the notes on the accompanying sheet.

2. [ ] The applicant is hereby notified that no International Search Report will be established and that the declaration under Article 17(2)(a) to that effect is transmitted herewith.

3. [ ] **With regard to the protest** against payment of (an) additional fee(s) under Rule 40.2, the applicant is notified that:

[ ] the protest together with the decision thereon has been transmitted to the International Bureau together with the applicant's request to forward the texts of both the protest and the decision thereon to the designated Offices.

[ ] no decision has been made yet on the protest; the applicant will be notified as soon as a decision is made.

4. **Further action(s):** The applicant is reminded of the following:

Shortly after **18 months** from the priority date, the international application will be published by the International Bureau. If the applicant wishes to avoid or postpone publication, a notice of withdrawal of the international application, or of the priority claim, must reach the International Bureau as provided in Rules 90*bis*.1 and 90*bis*.3, respectively, before the completion of the technical preparations for international publication.

Within **19 months** from the priority date, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until 30 months from the priority date (in some Offices even later).

Within **20 months** from the priority date, the applicant must perform the prescribed acts for entry into the national phase before all designated Offices which have not been elected in the demand or in a later election within 19 months from the priority date or could not be elected because they are not bound by Chapter II.

Name and mailing address of the International Searching Authority European Patent Office, P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Authorized officer Isabelle Porrachia

Form PCT/ISA/220 (July 1998)

NOTES TO FORM PCT/ISA/220

These Notes are intended to give the basic instructions concerning the filing of amendments under article 19. The Notes are based on the requirements of the Patent Cooperation Treaty, the Regulations and the Administrative Instructions under that Treaty. In case of discrepancy between these Notes and those requirements, the latter are applicable. For more detailed information, see also the PCT Applicant's Guide, a publication of WIPO.

In these Notes, "Article", "Rule", and "Section" refer to the provisions of the PCT, the PCT Regulations and the PCT Administrative Instructions respectively.

INSTRUCTIONS CONCERNING AMENDMENTS UNDER ARTICLE 19

The applicant has, after having received the international search report, one opportunity to amend the claims of the international application. It should however be emphasized that, since all parts of the international application (claims, description and drawings) may be amended during the international preliminary examination procedure, there is usually no need to file amendments of the claims under Article 19 except where, e.g. the applicant wants the latter to be published for the purposes of provisional protection or has another reason for amending the claims before international pbulication. Furthermore, it should be emphasized that provisional protection is available in some States only.

What parts of the international application may be amended?

Under Article 19, only the claims may be amended.

During the international phase, the claims may also be amended (or further amended) under Article 34 before the International Preliminary Examining Authority. The description and drawings may only be amended under Article 34 before the International Examining Authority.

Upon entry into the national phase, all parts of the international application may be amended under Article 28 or, where applicable, Article 41.

When?

Within 2 months from the date of transmittal of the international search report or 16 months from the priority date, whichever time limit expires later. It should be noted, however, that the amendments will be considered as having been received on time if they are received by the International Bureau after the expiration of the applicable time limit but before the completion of the technical preparations for international publication (Rule 46.1).

Where not to file the amendments?

The amendments may only be filed with the International Bureau and not with the receiving Office or the International Searching Authority (Rule 46.2).

Where a demand for international preliminary examination has been/is filed, see below.

How?

Either by cancelling one or more entire claims, by adding one or more new claims or by amending the text of one or more of the claims as filed.

A replacement sheet must be submitted for each sheet of the claims which, on account of an amendment or amendments, differs from the sheet originally filed.

All the claims appearing on a replacement sheet must be numbered in Arabic numerals. Where a claim is cancelled, no renumbering of the other claims is required. In all cases where claims are renumbered, they must be renumbered consecutively (Administrative Instructions, Section 205(b)).

The amendments must be made in the language in which the international application is to be published.

What documents must/may accompany the amendments?

Letter (Section 205(b)):

The amendments must be submitted with a letter.

The letter will not be published with the international application and the amended claims. It should not be confused with the "Statement under Article 19(1)" (see below, under "Statement under Article 19(1)").

The letter must be in English or French, at the choice of the applicant. However, if the language of the international application is English, the letter must be in English; if the language of the international application is French, the letter must be in French.

The letter must indicate the differences between the claims as filed and the claims as amended. It must, in particular, indicate, in connection with each claim appearing in the international application (it being understood that identical indications concerning several claims may be grouped),whether

(i) the claim is unchanged;

(ii) the claim is cancelled;

(iii) the claim is new;

(iv) the claim replaces one or more claims as filed;

(v) the claim is the result of the division of a claim as filed.

The following examples illustrate the manner in which amendments must be explained in the accompanying letter:

1. [Where originally there were 48 claims and after amendment of some claims there are 51]:
"Claims 1 to 29, 31, 32, 34, 35, 37 to 48 replaced by amended claims bearing the same numbers; claims 30, 33 and 36 unchanged; new claims 49 to 51 added."

2. [Where originally there were 15 claims and after amendment of all claims there are 11]:
"Claims 1 to 15 replaced by amended claims 1 to 11."

3. [Where originally there were 14 claims and the amendments consist in cancelling some claims and in adding new claims]:
"Claims 1 to 6 and 14 unchanged; claims 7 to 13 cancelled; new claims 15, 16 and 17 added." or
"Claims 7 to 13 cancelled; new claims 15, 16 and 17 added; all other claims unchanged."

4. [Where various kinds of amendments are made]:
"Claims 1-10 unchanged; claims 11 to 13, 18 and 19 cancelled; claims 14, 15 and 16 replaced by amended claim 14; claim 17 subdivided into amended claims 15, 16 and 17; new claims 20 and 21 added."

"Statement under article 19(1)" (Rule 46.4)

The amendments may be accompanied by a statement explaining the amendments and indicating any impact that such amendments might have on the description and the drawings (which cannot be amended under Article 19(1)).

The statement will be published with the international application and the amended claims.

It must be in the language in which the international appplication is to be published.

It must be brief, not exceeding 500 words if in English or if translated into English.

It should not be confused with and does not replace the letter indicating the differences between the claims as filed and as amended. It must be filed on a separate sheet and must be identified as such by a heading, preferably by using the words "Statement under Article 19(1)."

It may not contain any disparaging comments on the international search report or the relevance of citations contained in that report. Reference to citations, relevant to a given claim, contained in the international search report may be made only in connection with an amendment of that claim.

Consequence if a demand for international preliminary examination has already been filed

If, at the time of filing any amendments under Article 19, a demand for international preliminary examination has already been submitted, the applicant must preferably, at the same time of filing the amendments with the International Bureau, also file a copy of such amendments with the International Preliminary Examining Authority (see Rule 62.2(a), first sentence).

Consequence with regard to translation of the international application for entry into the national phase

The applicant's attention is drawn to the fact that, where upon entry into the national phase, a translation of the claims as amended under Article 19 may have to be furnished to the designated/elected Offices, instead of, or in addition to, the translation of the claims as filed.

For further details on the requirements of each designated/elected Office, see Volume II of the PCT Applicant's Guide.

PATENT COOPERATION TREATY

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference	**FOR FURTHER ACTION** see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
AJF57571/001		
International application No.	International filing date *(day/month/year)*	(Earliest) Priority Date *(day/month/year)*
PCT/GB 02/ 02488	28/05/2002	29/05/2001
Applicant		
THERMO FINNIGAN LLC		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 3 sheets.

[X] It is also accompanied by a copy of each prior art document cited in this report.

1. **Basis of the report**

   a. With regard to the **language,** the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

   [ ] the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

   b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing :

   [ ] contained in the international application in written form.

   [ ] filed together with the international application in computer readable form.

   [ ] furnished subsequently to this Authority in written form.

   [ ] furnished subsequently to this Authority in computer readble form.

   [ ] the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

   [ ] the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. [ ] **Certain claims were found unsearchable** (See Box I).

3. [ ] **Unity of invention is lacking** (see Box II).

4. With regard to the **title,**

   [X] the text is approved as submitted by the applicant.

   [ ] the text has been established by this Authority to read as follows:

5. With regard to the **abstract,**

   [X] the text is approved as submitted by the applicant.

   [ ] the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No. 1

   [ ] as suggested by the applicant.

   [X] because the applicant failed to suggest a figure.

   [ ] because this figure better characterizes the invention.

   [ ] None of the figures.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/GB 02/02488

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 H01J49/40

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 H01J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
EPO-Internal, WPI Data, PAJ, INSPEC

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X Y	US 5 898 173 A (FRANZEN JOCHEN) 27 April 1999 (1999-04-27) abstract ---	1,22 2,23
X	US 6 011 259 A (DRESCH THOMAS ET AL) 4 January 2000 (2000-01-04) abstract ---	1,22
Y	WO 99 67801 A (GONIN MARC ;IONWERKS (US)) 29 December 1999 (1999-12-29) cited in the application abstract ---	2,23
A	WO 98 21742 A (DAVIS LARRY J ;ROCKWOOD ALAN (US); SENSAR CORP (US)) 22 May 1998 (1998-05-22) cited in the application abstract --- -/--	1

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

° Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
3 December 2002	17/12/2002

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Hulne, S

1

Form PCT/ISA/210 (second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

International Application No
PCT/GB 02/02488

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication,where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 777 326 A (DAVIS LARRY J ET AL) 7 July 1998 (1998-07-07) cited in the application abstract	1
A	KRISTO M J ET AL: "SYSTEM FOR SIMULTANEOUS COUNT/CURRENT MEASUREMENT WITH A DUAL-MODE PHOTON/PARTICLE DETECTOR" REVIEW OF SCIENTIFIC INSTRUMENTS, AMERICAN INSTITUTE OF PHYSICS, US, vol. 59, no. 3, March 1988 (1988-03), pages 438-442, XP000103497 ISSN: 0034-6748 cited in the application abstract	1
A	US 6 229 142 B1 (COTTRELL JONATHAN C ET AL) 8 May 2001 (2001-05-08) cited in the application abstract	1

1

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
PCT/GB 02/02488

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
US 5898173 A	27-04-1999	DE	19635645 A1	05-03-1998
		GB	2317047 A ,B	11-03-1998
US 6011259 A	04-01-2000	AU	4149797 A	06-03-1998
		CA	2262627 A1	19-02-1998
		EP	0946267 A1	06-10-1999
		JP	2001500305 T	09-01-2001
		WO	9806481 A1	19-02-1998
WO 9967801 A	29-12-1999	AU	4580699 A	10-01-2000
		WO	9967801 A2	29-12-1999
WO 9821742 A	22-05-1998	US	5777326 A	07-07-1998
		US	6163032 A	19-12-2000
		US	6316768 B1	13-11-2001
		AU	7181898 A	03-06-1998
		EP	0939970 A1	08-09-1999
		JP	2001504265 T	27-03-2001
		WO	9821742 A1	22-05-1998
US 5777326 A	07-07-1998	AU	7181898 A	03-06-1998
		EP	0939970 A1	08-09-1999
		JP	2001504265 T	27-03-2001
		WO	9821742 A1	22-05-1998
US 6229142 B1	08-05-2001	EP	0970504 A2	12-01-2000
		EP	0970505 A2	12-01-2000
		EP	0970506 A2	12-01-2000
		WO	9938190 A2	29-07-1999
		WO	9938191 A2	29-07-1999
		WO	9938192 A2	29-07-1999
		JP	2001507513 T	05-06-2001
		JP	2001503196 T	06-03-2001
		JP	2000513494 T	10-10-2000
		US	6373052 B1	16-04-2002

PATENT COOPERATION TREATY

From the:
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

HILL, Richard.
WILSON GUNN M'CAW
41-51 Royal Exchange,
Cross street,
Manchester, M2 7BD
GRANDE BRETAGNE

PCT

WRITTEN OPINION

(PCT Rule 66)

Date of mailing *(day/month/year)*	24.08.2001
REPLY DUE	**within 2 month(s)** from the above date of mailing

Applicant's or agent's file reference		
RH/ALO/P/15339.WO		
International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/GB00/03332	31/08/2000	03/09/1999

International Patent Classification (IPC) or both national classification and IPC

H01J49/40

Applicant

MASSLAB LIMITED et al.

1. This written opinion is the **first** drawn up by this International Preliminary Examining Authority.

2. This opinion contains indications relating to the following items:

I	☒	Basis of the opinion
II	☐	Priority
III	☒	Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
IV	☐	Lack of unity of invention
V	☒	Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
VI	☐	Certain document cited
VII	☒	Certain defects in the international application
VIII	☒	Certain observations on the international application

24.10.01
DM

3. The applicant is hereby **invited to reply** to this opinion.

When? See the time limit indicated above. The applicant may, before the expiration of that time limit, request this Authority to grant an extension, see Rule 66.2(d).

How? By submitting a written reply, accompanied, where appropriate, by amendments, according to Rule 66.3. For the form and the language of the amendments, see Rules 66.8 and 66.9.

Also: For an additional opportunity to submit amendments, see Rule 66.4. For the examiner's obligation to consider amendments and/or arguments, see Rule 66.4 bis. For an informal communication with the examiner, see Rule 66.6.

If no reply is filed, the international preliminary examination report will be established on the basis of this opinion.

4. The final date by which the international preliminary examination report must be established according to Rule 69.2 is: 03/01/2002.

Name and mailing address of the international preliminary examining authority:
European Patent Office
D-80298 Munich
Tel. +49 89 2399 - 0 Tx: 523656 epmu d
Fax: +49 89 2399 - 4465

Authorized officer / Examiner
Lang, T

Formalities officer (incl. extension of time limits)
Schuster-Kaechele, W
Telephone No. +49 89 2399 2281

I. Basis of the opinion

1. With regard to the **elements** of the international application (Replacement *sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this opinion as "originally filed"*):

Description, pages:

1-12 as originally filed

Claims, No.:

1-14 as originally filed

Drawings, sheets:

1/7-7/7 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:

☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

III. Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1. The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been and will not be examined in respect of:

☐ the entire international application,

☒ claims Nos. 2, 14,

because:

☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (*specify*):

☒ the description, claims or drawings (*indicate particular elements below*) or said claims Nos. 2, 14 are so unclear that no meaningful opinion could be formed (*specify*):
see separate sheet

☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed.

☐ no international search report has been established for the said claims Nos. .

2. A written opinion cannot be drawn due to the failure of the nucleotide and/or amino acid sequence listing to comply with the standard provided for in Annex C of the Administrative Instructions:

☐ the written form has not been furnished or does not comply with the standard.

☐ the computer readable form has not been furnished or does not comply with the standard.

V. Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Claims	-
Inventive step (IS)	Claims	1, 3-12

Industrial applicability (IA) Claims -

2. Citations and explanations
see separate sheet

VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:
see separate sheet

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:
see separate sheet

Re Item III

Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1) The features of device **claim 2** relate only to a vague result to be achieved (attenuation of both incident ions and secondary electrons) by the use of the claimed device, which only amounts to stating the underlying problem. It is unclear by which device features this result could be achieved, contrary to Article 6 PCT. Even if claim 2 was a method claim directed to the use of a mass spectrometer, it would still be unclear by which features this result is to be achieved.

Since no clear determination of the scope of protection of this claim is possible, no judgement can be made on novelty or inventive step.

It appears, however, that the subject-matter of **claim 2** is not inventive for substantially the same reasons as given under point V for claim 1, since also the attenuation means of D1 or D3 are capable of attenuating both ions and secondary electrons.

2) Claim 14, in defining its subject-matter merely as being described with reference to the drawings, do not allow a clear determination of the scope of protection of the claim; and therefore no judgement can be made on novelty or inventive step.

According to Rule 6.2(a) PCT, claims should not contain such references except where absolutely necessary, which is not the case here.

It appears, however, that the subject-matter of **claim 14** is not inventive for substantially the same reasons as given under point V for claim 1.

Re Item V

Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1) Reference in made to the following documents:

D1: WO 99 38190 A (BATEMAN ROBERT HAROLD ;COTTRELL JONATHAN CHARLES (GB); GILBERT ANT) 29 July 1999 (1999-07-29)

D2: US 5463219 A; cited in D1

D3: MJ Kristo, CG Enke, System for simultaneous count/current measurement with a dual-mode photon/particle detector, Review of Scientific Instruments, vol 59(3), pages 438-442, 1988, XP001013497; cited in D1

Copies of D2 and D3 are attached.

2) Inventive Step (Article 33(3) PCT)

2.1) The document **D1**, which is considered to represent the most relevant state of the art, discloses (see abstract, Fig. 4 and pages 15-16) a time of flight mass spectrometer including two plate-shaped detector anodes 46, 44 arranged in series in the direction of ion travel, the first 46 attenuating the quantity of ions reaching the second 44, both connected to discriminators 28, 29 (which are TDCs, see the explanation in D1 page 2 lines 3 et seq., and in the present application).

From this, the subject-matter of **claim 1** differs only in that one of the detectors is additionally connected to an ADC, in parallel to the connection to the TDC.

This feature is not described as being part of the invention in the summary corresponding to claim 1 on page 3 of the description. No advantage or surprising effect of this feature can be derived from the application as filed, contrary to rule 5.1a) (iii) PCT. (Moreover, any later inclusion of such an effect would not comply with Art. 34 (2) b) PCT.)

The introductory portion of D1 (page 5 lines 17-20), and document **D2** cited there, disclose this feature as a possibility to extend the dynamic range of the detection system. Thus this feature is merely one of several straightforward possibilities from which the skilled person would select, in accordance with circumstances, in order to extend the dynamic range, thereby arriving at the subject-matter of claim 1 without the exercise of inventive skill.

2.2) For the same reasons, the subject-matter of claim 1 lacks an inventive step over

the prior art **D3** (see abstract, Fig. 2-4 and related text, in particular introduction on page 438), described in D1 from page 4 line 24 to page 5 line 17 (the intermediate electrode is a first detector and attenuation means; the electrode receiving electrons from the second multiplier is a second detector; the protection grid is an additional attenuation means).

A simultaneous detection as described in D1, page 5 lines 17-20, and in document **D2**, is again an obvious design option, readily included into one of the detectors of D3.

2.3) Dependent **claims 3-12** only comprise subject-matter relating to features which are either known, explicitly or implicitly, from the citations or considered to be routine matter to be expected of the skilled person. Therefore these claims cannot serve as a basis for a new independent claim which would meet the requirements of the PCT as to novelty and/or inventive step.

In particular, the subject-matter of **claims 3, 5, 6, 8-10, 11** (deceleration grid; see D1 page 4 line 38 et seq.; Fig. 4 item 46)**, and 12** is also disclosed in D1 as cited above.

(Pre-) amplifiers within discriminators (**claim 4**) or Faraday shields between detectors (**claim 7**) are well known standard features which do not provide any unexpected effect.

Re Item VII
Certain defects in the international application

1) It is left to the applicant to file new claims which take into account of the objections in this communication.

At the present understanding of the application, the subject-matter of claim 13 might possibly meet the requirements of Articles 33(2) and (3) PCT.

The applicant should also indicate in the letter of reply the difference of the subject-matter of the new independent claim vis-à-vis the state of the art (**reasons for novelty**) and the significance thereof (**reasons for inventive step**).

2) Independent claim 1 is not formulated in the two-part form in accordance with Rule 6.3(b)(i-ii) PCT such that features known in combination from the prior art are placed in the preamble of these claims.

If the applicant is of the opinion that a two-part form of the claim would be inappropriate he is invited to provide reasons in his reply.

3) The features of the claims are not provided with reference signs placed in parentheses (Rule 6.2(b) PCT).

4) The summary of the invention on page 3 lines 6-13 of the description is not in conformity with claim 1 as required by Rule 5.1(a)(iii) PCT.

5) The relevant background art disclosed in the documents **D1, D2,** and **D3** is not mentioned in the description, nor are these documents identified therein (Rule 5.1(a)(ii) PCT).

See PCT International Preliminary Examination Guidelines II-4.4.

The applicant should ensure that it is clear from the description which features of the subject-matter of the independent claim(s) are known from the prior art.

6) In order to facilitate the examination of the conformity of the amended application with the requirements of Article 34(2)(b) PCT, the applicant is requested to clearly identify the amendments carried out, no matter whether they concern amendments by addition, replacement or deletion, and to indicate the passages of the application as filed on which these amendments are based (Rule 66.8(a) PCT).

The applicant is asked to file amendments by way of replacement pages in the manner stipulated by Rule 66.8(a) PCT. In particular, fair copies of the amendments should be filed preferably in triplicate.

Moreover, the applicant's attention is drawn to the fact that, as a consequence of Rule 66.8(a) PCT the examiner is not permitted to carry out any amendments under the PCT procedure, however minor these may be.

Re Item VIII

Certain observations on the international application

(see also item I)

1) Claim 9 does not meet the requirements of Article 6 PCT in that the matter for which protection is sought is not clearly defined. The claim attempts to define the subject-matter in terms of the result to be achieved ("adapted to ...") which merely amounts to a statement of the underlying problem. The technical features necessary for achieving this result should be added (see e.g. claim 10).

2) The term "approximately" used in claim 13 is vague and unclear and leaves the reader in doubt as to the meaning of the technical feature to which it refers, thereby rendering the definition of the subject-matter of said claim unclear (Article 6 PCT).

Further, the term "mass" in claim 13 appears to stand for "mass spectrometer".

The Patent Office

INVESTOR IN PEOPLE

Thermo Masslab Limited
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Mr Cross/AJF
Received GC 21/02
Entered
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†**E-mail:** martyn.dixon@patent.gov.uk
Switchboard: 01633 814000
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DX 722540/41 Cleppa Park 3
http://www.patent.gov.uk

Your Reference: AJF/P57571/000
Application No: GB 0112963.4

19 February 2003

Dear Sirs

Patents Act 1977: Search Report under Section 17(5)

I enclose two copies of my search report and two copies of the citations.

DO YOU REQUIRE A REMINDER?

Publication

I estimate that, provided you have met all formal requirements, preparations for publication of your application will be completed soon after **25 March 2003**. You will then receive a letter informing you of completion and telling you the publication number and date of publication.

Amendment/withdrawal

If you wish to file amended claims for inclusion with the published application, or to withdraw the application to prevent publication, you must do so before the preparations for publication are completed. **No reminder will be issued.** If you write to the Office less than 3 weeks before the above completion date, please mark your letter prominently: **"URGENT - PUBLICATION IMMINENT"**.

Yours faithfully

Martyn Dixon
Examiner

RECEIVED
21 FEB 2003
BOULT WADE TENNANT

†**Use of E-mail:** Please note that e-mail should be used for correspondence only.

Application No: GB 0112963.4
Claims searched: all

Examiner: Martyn Dixon
Date of search: 18 February 2003

Patents Act 1977 : Search Report under Section 17

Documents considered to be relevant:

Category	Relevant to claims	Identity of document and passage or figure of particular relevance	
X	1,3-6, 20-22	GB 2246468 A	(Finnigan Mat) see e.g. fig 1 and page 7, line 3 to page 8, line 1
X	1,2,20	GB 1147667 A	(Friedrich-Schiller-Universitat) see figs 1 and 2
X	1-4,7,20	GB 0907511 A	(UKAEA) see fig 2 and page 2, line 100 to page 3, line 10
X	1-6,17-23	US 5026988 A	(Vanderbilt University) see fig 2 and col 4, line 40 to col 5, line 12

Categories:

X	Document indicating lack of novelty or inventive step	A	Document indicating technological background and/or state of the art.
Y	Document indicating lack of inventive step if combined with one or more other documents of same category.	P	Document published on or after the declared priority date but before the filing date of this invention.
&	Member of the same patent family	E	Patent document published on or after, but with priority date earlier than, the filing date of this application.

Field of Search:

Search of GB, EP, WO & US patent documents classified in the following areas of the UKC[V]:

H1D

Worldwide search of patent documents classified in the following areas of the IPC[7]:

H01J

The following online and other databases have been used in the preparation of this search report:

online: EPODOC,WPI,JAPIO,INSPEC

PATENT COOPERATION TREATY

PCT

4

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference. RH/ALO/P/15339.WO	**FOR FURTHER ACTION**	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
International application No. PCT/GB00/03332	International filing date *(day/month/year)* 31/08/2000	Priority date *(day/month/year)* 03/09/1999
International Patent Classification (IPC) or national classification and IPC H01J49/40		
Applicant MASSLAB LIMITED et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 8 sheets, including this cover sheet.

   ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of 7 sheets.

3. This report contains indications relating to the following items:

   - I ☒ Basis of the report
   - II ☐ Priority
   - III ☒ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - IV ☐ Lack of unity of invention
   - V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations suporting such statement
   - VI ☐ Certain documents cited
   - VII ☒ Certain defects in the international application
   - VIII ☒ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
03/04/2001	10.12.2001
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Lang, T Telephone No. +49 89 2399 2594

Form PCT/IPEA/409 (cover sheet) (January 1994)

☐ the description, pages:

☐ the claims, Nos.:

☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

III. Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1. The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:

☐ the entire international application.

☒ claims Nos. 2.

because:

☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (*specify*):

☒ the description, claims or drawings (*indicate particular elements below*) or said claims Nos. 2 are so unclear that no meaningful opinion could be formed (*specify*):
see separate sheet

☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed.

☐ no international search report has been established for the said claims Nos. .

2. A meaningful international preliminary examination cannot be carried out due to the failure of the nucleotide and/or amino acid sequence listing to comply with the standard provided for in Annex C of the Administrative Instructions:

☐ the written form has not been furnished or does not comply with the standard.

☐ the computer readable form has not been furnished or does not comply with the standard.

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	1, 3-13
	No:	Claims	-
Inventive step (IS)	Yes:	Claims	13
	No:	Claims	1, 3-12
Industrial applicability (IA)	Yes:	Claims	1, 3-13
	No:	Claims	-

2. Citations and explanations
see separate sheet

VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:
see separate sheet

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:
see separate sheet

Re Item III

Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1) The features of device **claim 2** relate only to a vague result to be achieved (attenuation of both incident ions and secondary electrons) by the use of the claimed device, which only amounts to stating the underlying problem. It is unclear by which device features this result could be achieved, contrary to Article 6 PCT. Even if claim 2 was a method claim directed to the use of a mass spectrometer, it would still be unclear by which features this result is to be achieved.

Since no clear determination of the scope of protection of this claim is possible, no judgement can be made on novelty or inventive step.

It appears, however, that the subject-matter of **claim 2** is not inventive for substantially the same reasons as given under point V for claim 1, since also the attenuation means of D1 or D3 are capable of attenuating both ions (by the cross-section of these means which intercepts the ion flux) and secondary electrons.

Re Item V

Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1) Reference in made to the following documents:

D1: WO 99 38190 A (BATEMAN ROBERT HAROLD ;COTTRELL JONATHAN CHARLES (GB); GILBERT ANT) 29 July 1999 (1999-07-29)

D2: US 5463219 A; cited in D1

D3: MJ Kristo, CG Enke, System for simultaneous count/current measurement with a dual-mode photon/particle detector, Review of Scientific Instruments, vol 59(3), pages 438-442, 1988, XP001013497; cited in D1

2) Inventive Step (Article 33(3) PCT)

2.1) The document **D1**, which is considered to represent the most relevant state of the art, discloses (see abstract, Fig. 4 and pages 15-16) a time of flight mass spectrometer including two plate-shaped detector anodes 46, 44 arranged in series in the direction of ion travel 33, the first 46 attenuating the quantity of ions reaching the second 44 (necessarily because of the cross-section of anode 46 of about 50% of the total area exposed to the ion flux 33). Both anodes are connected to discriminators 28, 29 (which *are* TDCs, see the explanation in D1 page 2 lines 3 et seq., and in the present application).

From this, the subject-matter of **claim 1** differs only in that one of the detectors is additionally connected to an ADC, in parallel to the connection to the TDC.

No advantage or surprising effect of this feature can be derived from the application as filed, contrary to rule 5.1a) (iii) PCT. (Moreover, any later inclusion of such an effect would not comply with Art. 34 (2) b) PCT.)

The introductory portion of D1 (page 5 lines 17-20), and document **D2** cited there, disclose this feature as a possibility to extend the dynamic range of the detection system. Thus this feature is merely one of several straightforward possibilities from which the skilled person would select, in accordance with circumstances, in order to extend the dynamic range, thereby arriving at the subject-matter of claim 1 without the exercise of inventive skill.

2.2) For the same reasons, the subject-matter of claim 1 lacks an inventive step over the prior art **D3** (see abstract, Fig. 2-4 and related text, in particular introduction on page 438), described in D1 from page 4 line 24 to page 5 line 17 (the intermediate electrode is a first detector and attenuation means (since it intercepts the ion flux); the electrode receiving electrons from the second multiplier is a second detector. The protection grid also intercepts the ion flux and is an additional attenuation means).

A simultaneous detection as described in D1, page 5 lines 17-20, and in document **D2**, is again an obvious design option, readily included into one of the detectors of D3.

2.3) Dependent **claims 3-11** only comprise subject-matter relating to features which are either known, explicitly or implicitly, from the citations or considered to be routine matter to be expected of the skilled person. Therefore these claims cannot serve as a basis for a new independent claim which would meet the requirements of the PCT as to novelty and/or inventive step.

In particular, the subject-matter of **claims 3, 5, 6, 8** (interception and thus attenuation of the ion flux by one of the detector electrodes is necessarily also present in D1 and D3), **9, 10** (deceleration grid; see D1 page 4 line 38 et seq.; Fig. 4 item 46), **and 11** is also disclosed in D1 or D3 as cited above.

(Pre-) amplifiers within discriminators (**claim 4**) or Faraday shields between detectors (**claim 7**) are well known standard features which do not provide any unexpected effect.

2.4) The cross-sectional area of the perforations as claimed in **claim 12** is not suggested or rendered obvious in the available prior art and may be regarded as solving the problem of allowing a much more quantitative analysis of the incoming ion beam (page 8 line 20 of the application), thus meeting the requirements of Article 33(3) PCT.

Re Item VII

Certain defects in the international application

1) Independent claim 1 is not formulated in the two-part form in accordance with Rule 6.3(b)(i-ii) PCT such that features known in combination from the prior art are placed in the preamble of these claims.

2) The features of the claims are not provided with reference signs placed in parentheses (Rule 6.2(b) PCT).

3) The relevant background art disclosed in the documents **D1, D2,** and **D3** is not mentioned in the description, nor are these documents identified therein (Rule 5.1(a)(ii) PCT). See PCT International Preliminary Examination Guidelines II-4.4.

Re Item VIII

Certain observations on the international application

(See also item III.)

The statement on page 10 lines 19-20 (that the examples of figures 2-5 were not part of the invention, although they apparently fall within the wording of claim 1) is in contradiction to page 6 line 16 of the description and to the subject-matter of **claim 8**, thereby resulting in lack of clarity of the claims (Article 6 PCT) when used to interpret them.

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB00/03332

I. Basis of the report

1. With regard to the **elements** of the international application *(Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17))*:

Description, pages:

1,2,6-9,11,12	as originally filed			
3-5,10	as received on	23/11/2001	with letter of	23/11/2001

Claims, No.:

1-12	as received on	23/11/2001	with letter of	23/11/2001

Drawings, sheets:

1/7-7/7	as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

   These elements were available or furnished to this Authority in the following language: , which is:

   - ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
   - ☐ the language of publication of the international application (under Rule 48.3(b)).
   - ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

   - ☐ contained in the international application in written form.
   - ☐ filed together with the international application in computer readable form.
   - ☐ furnished subsequently to this Authority in written form.
   - ☐ furnished subsequently to this Authority in computer readable form.
   - ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
   - ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
RH/ALO/P/15339.WO		
International application No.	International filing date *(day/month/year)*	Priority date *(day/month/year)*
PCT/GB00/03332	31/08/2000	03/09/1999
International Patent Classification (IPC) or national classification and IPC H01J49/40		
Applicant MASSLAB LIMITED et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 8 sheets, including this cover sheet.

   ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of 7 sheets.

3. This report contains indications relating to the following items:

   - I ☒ Basis of the report
   - II ☐ Priority
   - III ☒ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - IV ☐ Lack of unity of invention
   - V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations suporting such statement
   - VI ☐ Certain documents cited
   - VII ☒ Certain defects in the international application
   - VIII ☒ Certain observations on the International application

Date of submission of the demand	Date of completion of this report
03/04/2001	10.12.2001
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Lang, T Telephone No. +49 89 2399 2594

Form PCT/IPEA/409 (cover sheet) (January 1994)

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB00/03332

I. Basis of the report

1. With regard to the elements of the international application *(Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17))*:

Description, pages:

1,2,6-9,11,12	as originally filed			
3-5,10	as received on	23/11/2001	with letter of	23/11/2001

Claims, No.:

1-12	as received on	23/11/2001	with letter of	23/11/2001

Drawings, sheets:

1/7-7/7	as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

   These elements were available or furnished to this Authority in the following language: , which is:

   - ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
   - ☐ the language of publication of the international application (under Rule 48.3(b)).
   - ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

   - ☐ contained in the international application in written form.
   - ☐ filed together with the international application in computer readable form.
   - ☐ furnished subsequently to this Authority in written form.
   - ☐ furnished subsequently to this Authority in computer readable form.
   - ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
   - ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB00/03332

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

   (Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

III. Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1. The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:

   ☐ the entire international application.

   ☒ claims Nos. 2.

because:

   ☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (*specify*):

   ☒ the description, claims or drawings (*indicate particular elements below*) or said claims Nos. 2 are so unclear that no meaningful opinion could be formed (*specify*):
   see separate sheet

   ☐ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed.

   ☐ no international search report has been established for the said claims Nos. .

2. A meaningful international preliminary examination cannot be carried out due to the failure of the nucleotide and/or amino acid sequence listing to comply with the standard provided for in Annex C of the Administrative Instructions:

   ☐ the written form has not been furnished or does not comply with the standard.

   ☐ the computer readable form has not been furnished or does not comply with the standard.

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB00/03332

1. Statement

Novelty (N)	Yes:	Claims	1, 3-13
	No:	Claims	-
Inventive step (IS)	Yes:	Claims	13
	No:	Claims	1, 3-12
Industrial applicability (IA)	Yes:	Claims	1, 3-13
	No:	Claims	-

2. Citations and explanations
see separate sheet

VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:
see separate sheet

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:
see separate sheet

INTERNATIONAL PRELIMINARY EXAMINATION REPORT - SEPARATE SHEET

International application No. PCT/GB00/03332

Re Item III

Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1) The features of device **claim 2** relate only to a vague result to be achieved (attenuation of both incident ions and secondary electrons) by the use of the claimed device, which only amounts to stating the underlying problem. It is unclear by which device features this result could be achieved, contrary to Article 6 PCT. Even if claim 2 was a method claim directed to the use of a mass spectrometer, it would still be unclear by which features this result is to be achieved.

Since no clear determination of the scope of protection of this claim is possible, no judgement can be made on novelty or inventive step.

It appears, however, that the subject-matter of **claim 2** is not inventive for substantially the same reasons as given under point V for claim 1, since also the attenuation means of D1 or D3 are capable of attenuating both ions (by the cross-section of these means which intercepts the ion flux) and secondary electrons.

Re Item V

Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1) Reference in made to the following documents:

D1: WO 99 38190 A (BATEMAN ROBERT HAROLD ;COTTRELL JONATHAN CHARLES (GB); GILBERT ANT) 29 July 1999 (1999-07-29)

D2: US 5463219 A; cited in D1

D3: MJ Kristo, CG Enke, System for simultaneous count/current measurement with a dual-mode photon/particle detector, Review of Scientific Instruments, vol 59(3), pages 438-442, 1988, XP001013497; cited in D1

2) Inventive Step (Article 33(3) PCT)

2.1) The document **D1**, which is considered to represent the most relevant state of the art, discloses (see abstract, Fig. 4 and pages 15-16) a time of flight mass spectrometer including two plate-shaped detector anodes 46, 44 arranged in series in the direction of ion travel 33, the first 46 attenuating the quantity of ions reaching the second 44 (necessarily because of the cross-section of anode 46 of about 50% of the total area exposed to the ion flux 33). Both anodes are connected to discriminators 28, 29 (which *are* TDCs, see the explanation in D1 page 2 lines 3 et seq., and in the present application).

From this, the subject-matter of **claim 1** differs only in that one of the detectors is additionally connected to an ADC, in parallel to the connection to the TDC.

No advantage or surprising effect of this feature can be derived from the application as filed, contrary to rule 5.1a) (iii) PCT. (Moreover, any later inclusion of such an effect would not comply with Art. 34 (2) b) PCT.)

The introductory portion of D1 (page 5 lines 17-20), and document **D2** cited there, disclose this feature as a possibility to extend the dynamic range of the detection system. Thus this feature is merely one of several straightforward possibilities from which the skilled person would select, in accordance with circumstances, in order to extend the dynamic range, thereby arriving at the subject-matter of claim 1 without the exercise of inventive skill.

2.2) For the same reasons, the subject-matter of claim 1 lacks an inventive step over the prior art **D3** (see abstract, Fig. 2-4 and related text, in particular introduction on page 438), described in D1 from page 4 line 24 to page 5 line 17 (the intermediate electrode is a first detector and attenuation means (since it intercepts the ion flux); the electrode receiving electrons from the second multiplier is a second detector. The protection grid also intercepts the ion flux and is an additional attenuation means).

A simultaneous detection as described in D1, page 5 lines 17-20, and in document **D2**, is again an obvious design option, readily included into one of the detectors of D3.

2.3) Dependent **claims 3-11** only comprise subject-matter relating to features which are either known, explicitly or implicitly, from the citations or considered to be routine matter to be expected of the skilled person. Therefore these claims cannot serve as a basis for a new independent claim which would meet the requirements of the PCT as to novelty and/or inventive step.

In particular, the subject-matter of **claims 3, 5, 6, 8** (interception and thus attenuation of the ion flux by one of the detector electrodes is necessarily also present in D1 and D3), **9, 10** (deceleration grid; see D1 page 4 line 38 et seq.; Fig. 4 item 46), **and 11** is also disclosed in D1 or D3 as cited above.

(Pre-) amplifiers within discriminators (**claim 4**) or Faraday shields between detectors (**claim 7**) are well known standard features which do not provide any unexpected effect.

2.4) The cross-sectional area of the perforations as claimed in **claim 12** is not suggested or rendered obvious in the available prior art and may be regarded as solving the problem of allowing a much more quantitative analysis of the incoming ion beam (page 8 line 20 of the application), thus meeting the requirements of Article 33(3) PCT.

Re Item VII

Certain defects in the international application

1) Independent claim 1 is not formulated in the two-part form in accordance with Rule 6.3(b)(i-ii) PCT such that features known in combination from the prior art are placed in the preamble of these claims.

2) The features of the claims are not provided with reference signs placed in parentheses (Rule 6.2(b) PCT).

3) The relevant background art disclosed in the documents **D1, D2,** and **D3** is not mentioned in the description, nor are these documents identified therein (Rule 5.1(a)(ii) PCT). See PCT International Preliminary Examination Guidelines II-4.4.

INTERNATIONAL PRELIMINARY EXAMINATION REPORT - SEPARATE SHEET

International application No. PCT/GB00/03332

Re Item VIII

Certain observations on the international application

(See also item III.)

The statement on page 10 lines 19-20 (that the examples of figures 2-5 were not part of the invention, although they apparently fall within the wording of claim 1) is in contradiction to page 6 line 16 of the description and to the subject-matter of **claim 8**, thereby resulting in lack of clarity of the claims (Article 6 PCT) when used to interpret them.